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UNITED STATES DEPARTMENT OF THE INTERIOR
Mining Enforcement and Safety Administration
Washington, D. C. 20240

U. S. Mining Enforcement and Safety Administration.

Informational Report 1015

Sampling Mine Atmospheres for Potential Alpha Energy Due to the Presence of Radon-220 (Thoron) Daughters

By R. L. Rock
Denver Technical Support Center, Denver, Colo.

UNITED STATES DEPARTMENT OF THE INTERIOR
Rogers C. B. Morton, Secretary

Mining Enforcement and Safety Administration
James M. Day, Administrator

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SAMPLING MINE ATMOSPHERES FOR POTENTIAL ALPHA ENERGY DUE TO THE PRESENCE OF RADON-220 (THORON) DAUGHTERS

by

R. L. Rock[1]

ABSTRACT

Although thorium minerals are not commonly found with uranium minerals mined in the United States, the mineral combination has been observed in a few deposits. The possibility also exists that thorium minerals might be associated with nonuranium ores or that thorium may be a constituent of the rocks surrounding nonradioactive ore deposits.

Thorium-232, like uranium-238, is the parent of a decay series which includes a dense radioactive gas. In the thorium series this gas (radon-220) is commonly called thoron. Thoron decays through a series of short-lived daughters, some of which are alpha-particle emitters, and can pose the same health hazard (lung cancer) as has been attributed to excessive exposure to radon daughters.

This paper describes the decay characteristics of thoron and its daughters, and gives a simple field method for testing for the presence of thoron daughters. Where such a test proves positive, the procedure for determining potential alpha energy (working levels) from thoron daughters is given.

The occurrence of thoron (radon-220) and radon (radon-222) daughters together complicates the problem of computing the working level attributable to each decay series. A system is given for determining if a mixture of thoron and radon daughters is present, and the method for computing the total potential alpha energy for such mixtures is described.

Throughout this paper, thoron daughters refers to daughters of the isotope radon-220 and radon daughters refers to daughters of the isotope radon-222.

INTRODUCTION

Minerals containing thorium-232 are quite common in many rocks, but the limited market for thorium metal has precluded extensive mining of thorium ores. The use of thorium as an alloy with other metals is becoming more

[1]Chief, Radiation Group.

popular, but important increases in future demands for thorium metal, depend greatly on its use as a fertile material for breeder reactors.

A field method for measuring airborne thoron-daughter concentrations was developed by the U.S. Public Health Service in 1960 (1).[2] Bureau personnel had an opportunity to apply this measurement method while characterizing the radiation hazards in an Alaskan high-grade uranium mine during 1971. The average grade of the ore was over 1.0 percent U_3O_8 and about 1.0 percent ThO_2 accompanied the ore. The presence of both thoron and radon daughters in the mine atmosphere necessitated special sampling and data-reduction procedures to allow measured alpha activity to be correctly interpreted in terms of total potential alpha energy. The basic problem was to devise a simple means of segregating the alpha activities of thoron and radon daughters which were collected simultaneously on a filter paper. The importance of performing this separation is emphasized by the fact that thoron daughter alpha activity denotes about 10 times as much potential alpha energy as an equal amount of radon-daughter alpha activity.

ACKNOWLEDGMENTS

The work of U.S. Public Health Service personnel in developing field methods for both radon- and thoron-daughter sampling has proven invaluable to health officials representing the mineral industries (1-2).

The Dawn Mining Company was most cooperative in inviting Bureau of Mines personnel to study the unusual radiation conditions in their Kendrick Bay, Alaska, mine. Much of the practical information presented herein, for rapidly identifying and evaluating the two main types of airborne radioactive contaminants occurring in mine atmospheres, was field-tested during the study of the Dawn Mining Company's Kendrick Bay, Alaska, uranium mine.

DECAY CHARACTERISTICS

Much has been written regarding the decay pattern of the uranium-238 series and, more particularly, about the radon and radon-daughter transient segment of this series (2-3). Because this paper is concerned primarily with distinguishing and evaluating radon-220 (thoron) daughters which could easily be confused with radon-222 daughters, an understanding of the differences in the decay patterns of the two series is important.

Radon Daughters

Figure 1 depicts the decay pattern for radon daughters commencing with the parent, radon gas.

Only four daughters of radon [RaA(Po-218), RaB(Pb-214), RaC(Bi-214), and RaC'(Po-214)] have significance so far as a rapid release of alpha energy is concerned. Of these, only Po-218 and Po-214 are actually alpha-particle

[2]Underlined numbers in parentheses refer to items in the list of references at the end of this report.

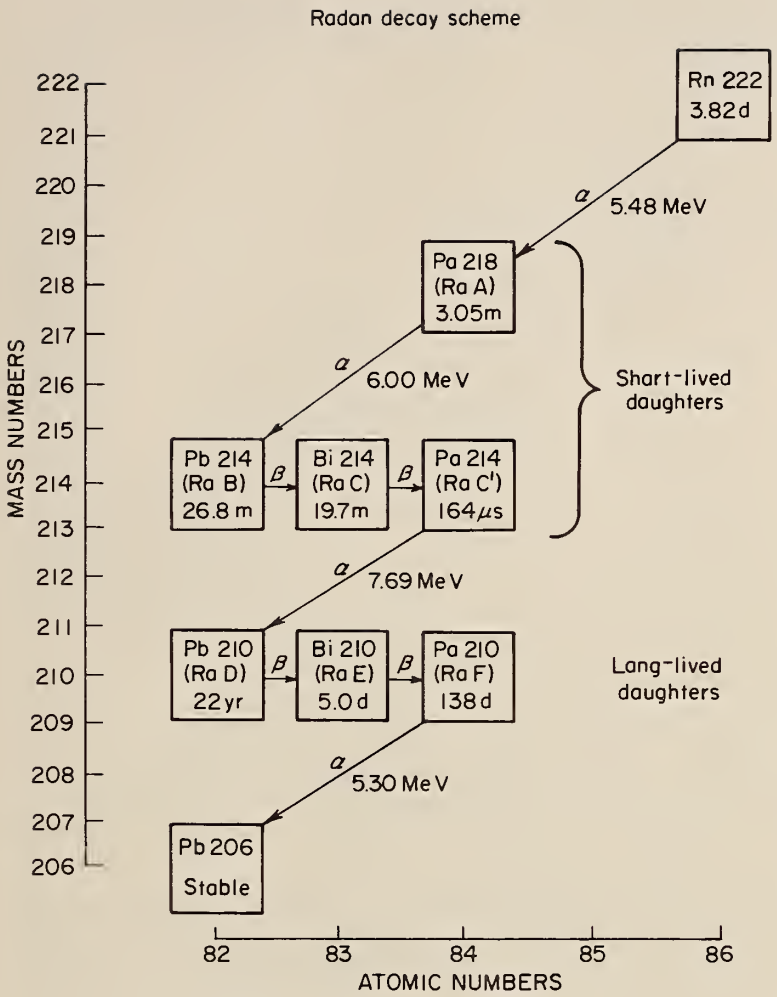

FIGURE 1. - Radon decay scheme.

emitters. Because of the short half-life of Po-214 (164μ seconds), Po-214 atoms are never present in mine air in sufficient abundance to contribute significant alpha energy to lung tissue. The importance of Po-214 is that each atom of Po-218, Pb-214, and Bi-214 trapped in the respiratory system decays through Po-214 to Pb-210 within a few hours, releasing a 7.7 MeV alpha particle in the process.

Decay beyond Pb-210 is not considered important as a health hazard because the alpha-energy release rate of Po-210 is slowed so drastically by the 22-year half-life of Pb-210.

The important feature in identifying radon daughters is their characteristic half-life which is about 30 minutes following the decay of Po-218. During the first 20 minutes following sampling, 99 percent of the Po-218 (3.05-minute half-life) will have decayed to Pb-214 (26.8-minute half-life). After the Po-218 has decayed, all alpha activity counted will be caused by the decay of Po-214 resulting from the decay of its precursors Pb-214 and Bi-214. A plot of the alpha decay curve for pure radon daughters (see fig. 2) will exhibit an average 30-minute half-life until Bi-214 has grown into equilibrium with Pb-214. The 27-minute half-life of Pb-214 will thereafter characterize the radon-daughter decay pattern.

Thoron Daughters

The complete thoron-daughter decay scheme is shown in figure 3. Thoron and its daughters, like radon daughters, are a transient series within a major radioactive series. The overall series is initiated by the decay of thorium-232.

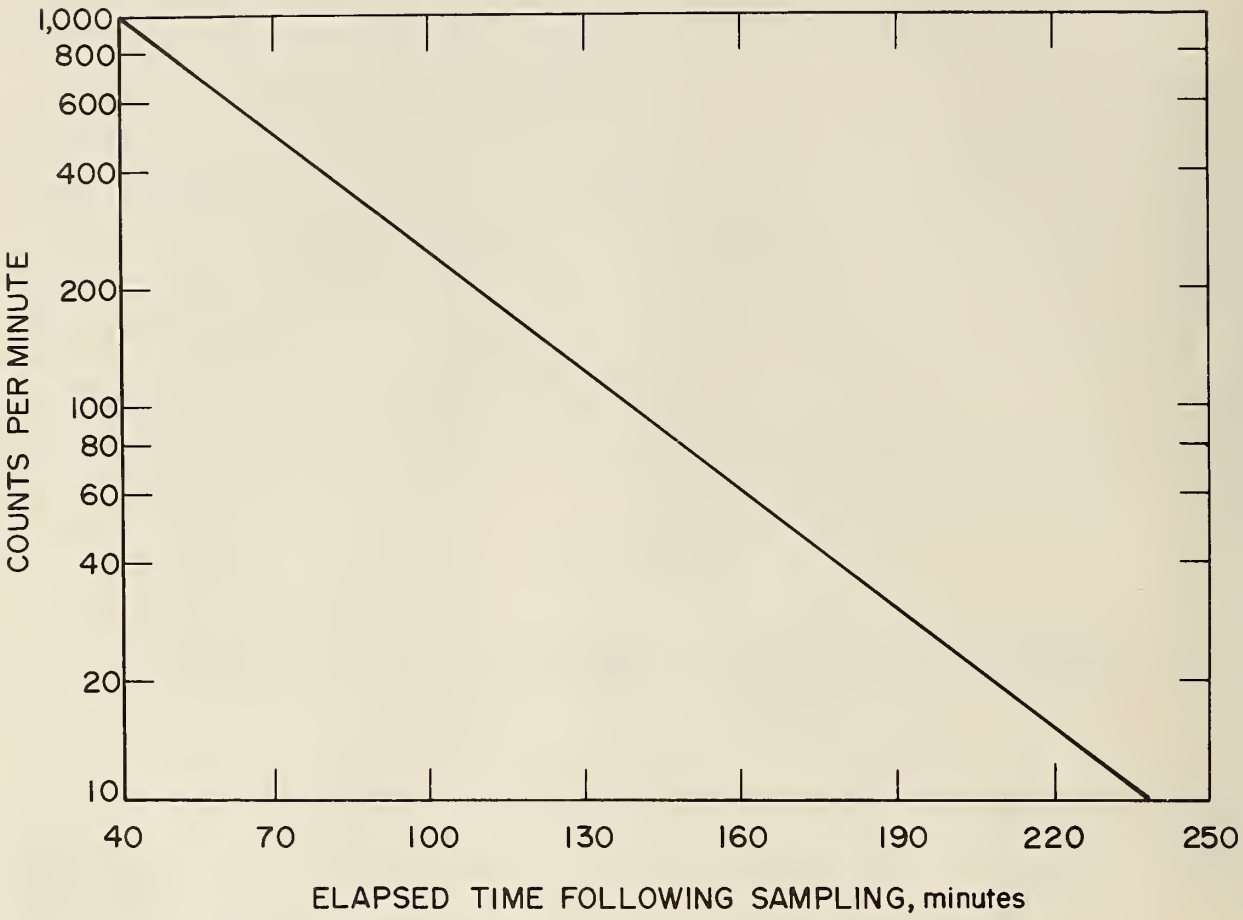

FIGURE 2. - Decay plotted against time for pure radon daughters.

Referring to figure 3, radon-220 (thoron), ThA (Po-216), ThC (Bi-212), and ThC' (Po-212) are all alpha-particle emitters. Thoron gas, like radon, can be ignored as a health hazard because it does not build up in the lungs. Po-216, with its very short half-life (0.16 sec), delivers an inconsequential radiation dose because so few atoms can accumulate. ThB, while not an alpha-particle emitter itself, has a long half-life (10.6 hours). This means that as thoron daughters approach equilibrium with thoron gas, the number of Pb-212 atoms present will be so much greater than the number of atoms of any of the other daughters, that only Pb-212 need be considered in evaluating potential alpha energy. Even Bi-212, with its 60-minute half-life, would contribute only an additional 8 percent of the alpha energy if it were initially in equilibrium with thoron (1). Under reasonably good ventilation conditions, the Bi-212 contribution due to direct inhalation would be only a few percent of the total dose delivered to the lungs.

Each atom of Pb-212 decays through Bi-212. Bi-212 atoms decay by a split process in which 33.7 percent release a 6.07 MeV alpha particle and decay to Tl-208. The other 66.3 percent of the Bi-212 atoms decay to Po-212 by releasing a beta particle. Po-212 atoms decay almost instantaneously by

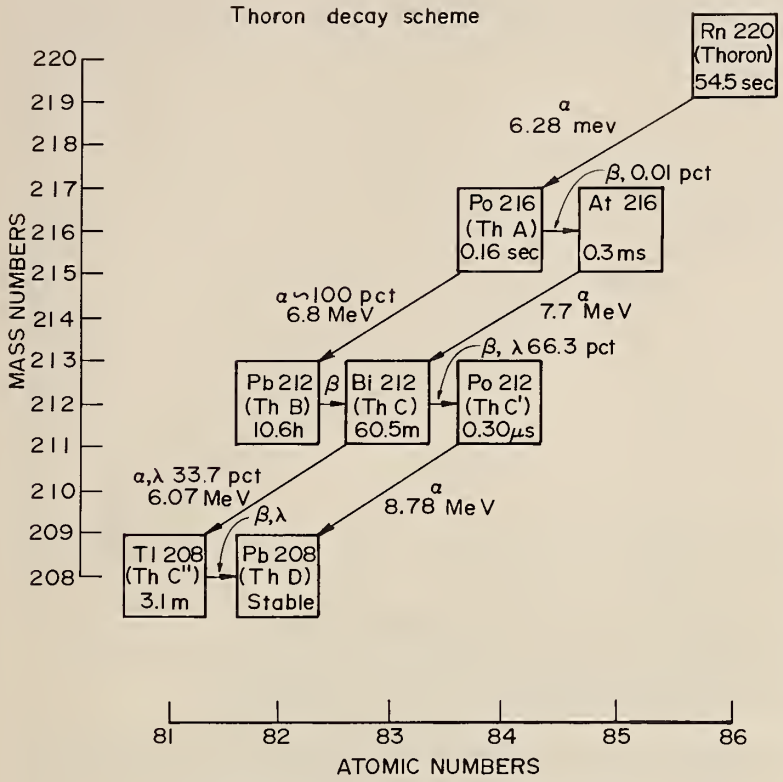

FIGURE 3. - Thoron decay scheme.

releasing an 8.78 MeV alpha particle and become stable Pb-208. For all practical purposes, the potential alpha energy from thoron daughters can be calculated by multiplying the number of Pb-212 atoms present by an average potential alpha energy of 7.86 MeV.

A plot of the alpha radiation decay rate for pure thoron daughters will reflect the 10.6-hour half-life of Pb-212 after Bi-212 has grown into equilibrium with Pb-212 on the sample. This typical decay curve is depicted by figure 4A. Bi-212 will never be in complete equilibrium in the mine atmosphere, but will grow into equilibrium on the filter paper sample within a matter of a few hours (usually 3-5 hours), depending upon the initial equilibrium relationship. Figure 4B shows a typical decay curve of Pb-212 and Bi-212 in disequilibrium during the first 3 hours following sampling.

RADON DAUGHTER-THORON DAUGHTER MIXTURES

Radon daughter-thoron daughter mixtures collected on a filter paper will demonstrate a different decay curve from either figure 2 or figures 4A and 4B.

In the early stages of decay, the presence of radon daughters on the sample will be depicted by a rapid reduction in activity with time, depending upon the initial number of Po-218, Pb-214, and Bi-214 atoms present in relation to the number of Pb-212 and Bi-212 atoms present. In a sample with nearly all activity due to Po-218, Pb-214, and Bi-214, the decay curve will dip sharply initially and gradually flatten as the radon daughters decay with a 30-minute half-life, leaving the much longer lived Pb-212 decaying at a slower rate. Figure 5 illustrates this situation.

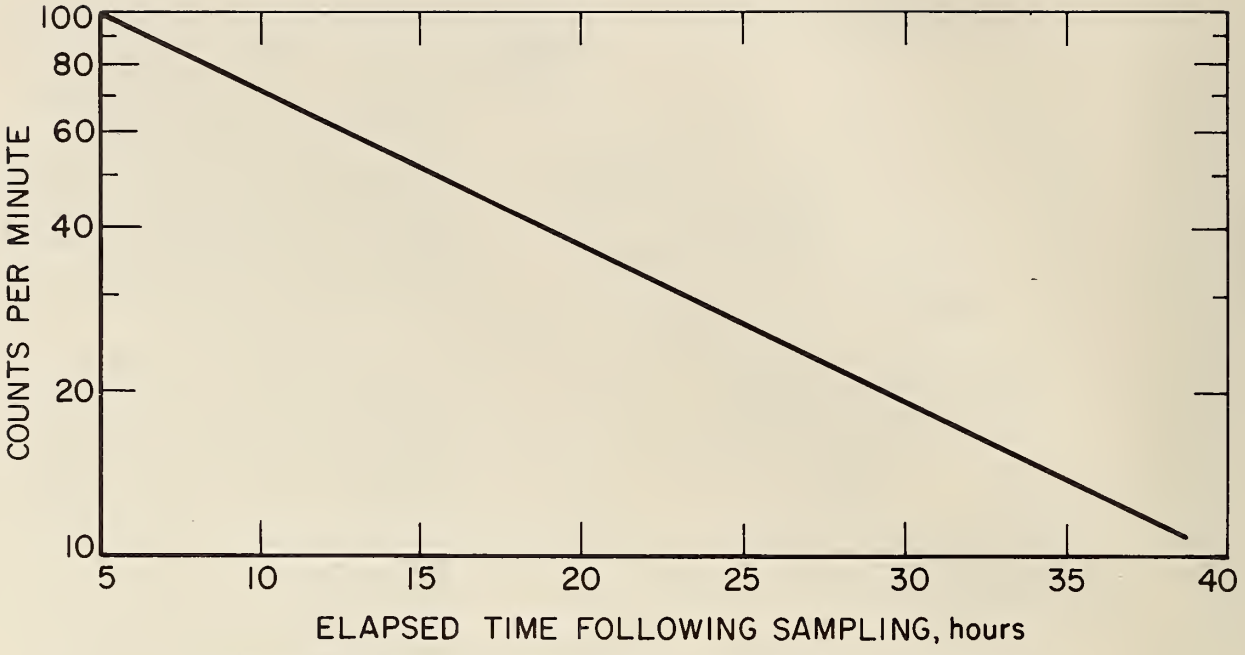

FIGURE 4A. - Decay plotted against time for pure thoron daughters.

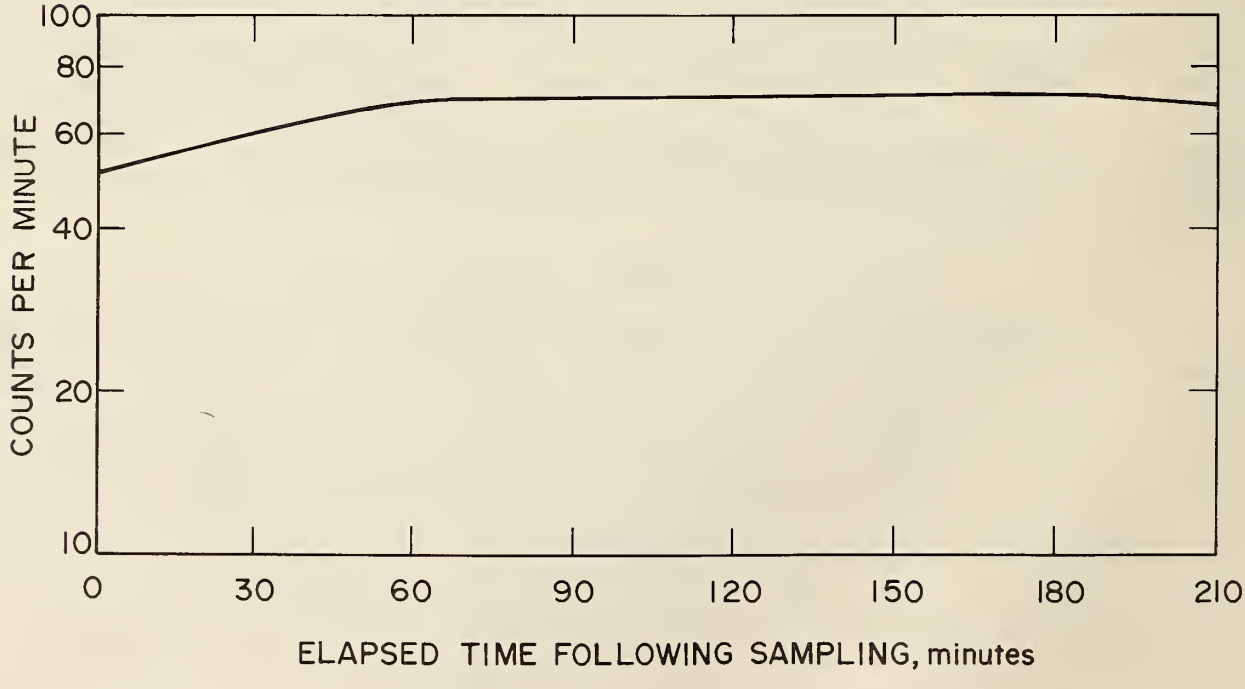

FIGURE 4B. - Decay plotted against time for pure thoron daughters at slight disequilibrium during early decay.

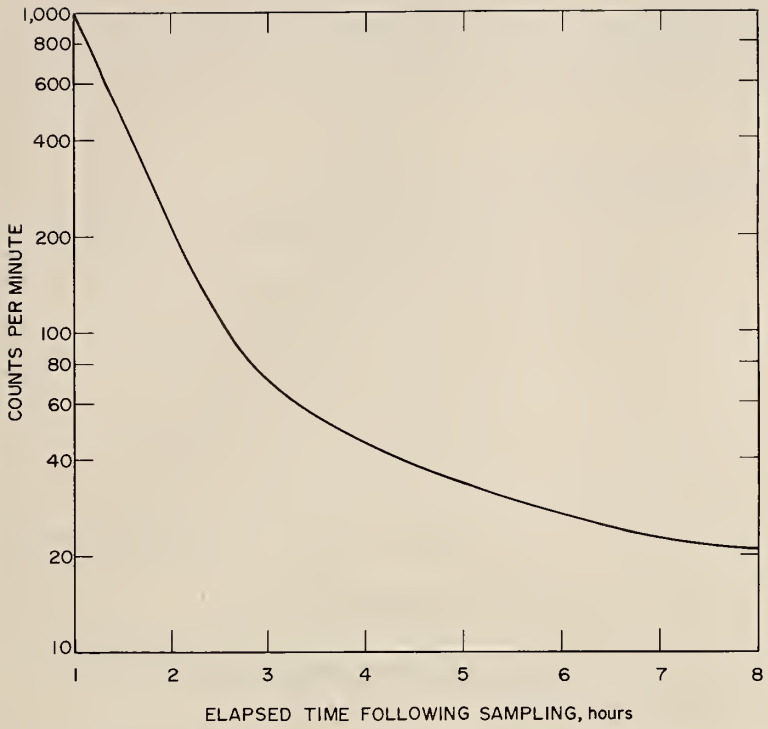

FIGURE 5. - Decay plotted against time for a hypothetical radon daughter-thoron daughter mixture (predominantly radon daughters).

At the other extreme, that is nearly all activity due to Pb-212 and Bi-212, there may be only a slight initial reduction or even an increase in the total activity after 30 minutes, and the decay curve will flatten much more rapidly, as illustrated by figure 6.

SAMPLING AND EVALUATION PROCEDURES

Screening Samples To Test for the Presence of Airborne Radioactive Particulates

General screening samples are recommended for all underground mines to test for the possible presence of airborne radioactive particulates. If such samples are not taken, a health hazard from radiation might remain undetected.

The screening sample should consist of at least 50 liters of air filtered through a suitable filter (fiberglass or membrane). This amount of collected aerosol from the air should allow sufficient sensitivity for thoron-daughter detection. A high-volume pump (10-1pm capacity) is convenient for this type of sampling because it allows the collection of the 50-liter sample in only 5 minutes.

Sample location selection is important because the hazard may not be detectable in all parts of the mine. If a radiation hazard due to airborne particulates exists, it is most likely to be evident in poorly ventilated workings and in return air. Therefore, screening samples should be taken in those areas.

Filter samples, taken for screening purposes, can be counted immediately to determine if any radioactivity is present. If radioactivity is not detectable, the test can be immediately considered negative. If radioactivity is detectable on the filter paper, the types of particulates responsible can be determined by their decay pattern.

Starting about 30 minutes after sampling, count the sample every 30 minutes for a total of about 4 counts. If the decay curve consistently represents about a 30-minute half-life, radon daughters are the radionuclides to

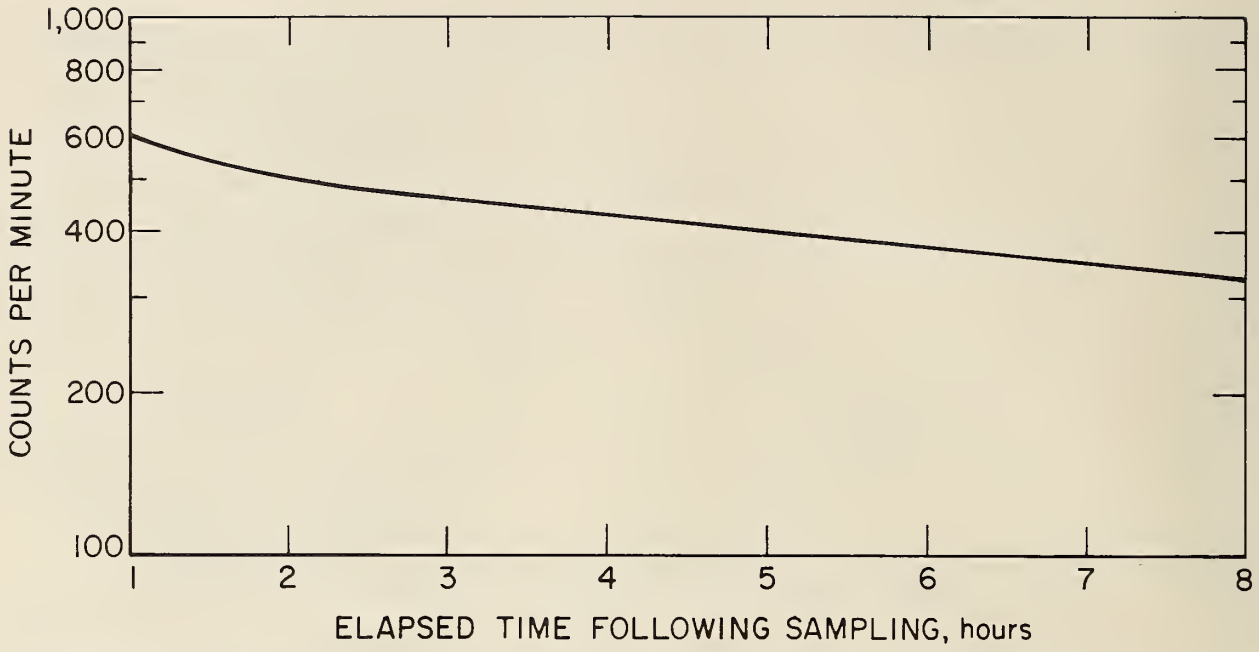

FIGURE 6. - Decay plotted against time for a hypothetical thoron daughter-radon daughter mixture (predominantly thoron daughters).

be evaluated. If decay indicates a half-life of more than 30 minutes, thoron daughters are probably present. The sample should then be counted at subsequent intervals until at least 5 hours have elapsed.

Where the decay rate decreases very slowly during the first counting intervals or increases slightly and then diminishes very slowly, thoron daughters can be considered to constitute the major portion of the radionuclides present.

Radon-Daughter Sampling

Radon-daughter sampling does not require so large an air sample volume as is required for thoron-daughter sampling, but the 50-liter or larger samples suggested for screening samples will allow greater radon-daughter sensitivity. The important thing to remember in this respect is that the total sample volume for radon daughters should be collected in a 5-minute period. Otherwise, correction factors may be necessary for the buildup and decay of the radionuclides being collected during sampling.

Alpha-particle activity on the filter paper is counted 40 to 90 minutes after sampling using an alpha scintillation detector coupled to a count-rate meter or a digital scaler. The efficiency of the detector for counting 7.7 MeV alpha particles due to the decay of Po-214 must be known. Thus, counts per minute (cpm) can be converted to the actual disintegrations per minute (dpm) occurring on the filter. Working levels (WL), the measure of potential alpha energy, can then be calculated by dividing the total disintegrations per minute by the liters of air sampled, multiplied by the appropriate factor for the elapsed time following sampling. These factors are shown in figure 7.

The technical definition of a working level of radon (radon-222) daughters is any combination of Po-218, Pb-214, and Bi-214 per liter or air which will yield 1.3×10^5/MeV of alpha energy in decaying completely through Po-214 to Pb-210.

Following is a sample radon-daughter WL calculation:

Sample volume = 10 liters

Time at end of sample = 10:15 a.m.

Time of counting = 11:05 a.m.

Elapsed time = 50 minutes

Time factor (from fig. 7) = 130

Count rate (cpm) = 250

Counter efficiency = 25 percent

Total decay events per minute = $\frac{250}{0.25}$ = 1,000 dpm

WL = $\frac{1,000}{10 \times 130}$ = 0.77

Thoron-Daughter Sampling

The high-volume screening sample, 50 liters, can be used directly for evaluating thoron daughters, if their presence is indicated by sample radioactive decay characteristics. Unlike radon daughters, buildup and decay of collected thoron daughters do not present a problem during a total sampling time of up to 1 hour. This is because of the 10.6-hour half-life of Pb-212, the radionuclide of importance.

For the thoron-daughter working level computation, the sample is allowed to age for at least 5 hours after sampling before counting. This elapsed time allows Bi-212 to grow into equilibrium with Pb-212 so that the combined alpha counts of Bi-212 and Po-212 may be directly correlated with the number of atoms of Pb-212 present on the filter. A secondary advantage is that any radon daughters which may have accompanied the thoron daughters will have decayed sufficiently to not interfere with the thoron-daughter working level computation.

Counts per minute on the filter paper are converted to total disintegrations per minute by dividing by the appropriate detector efficiency factor. Most scintillation detectors are not too energy dependent; therefore, the efficiency factor for counting Po-214 can usually be applied when counting the combined 6.07 MeV alpha particles and 8.78 MeV alpha particles from Bi-212 and Po-212, respectively. Detector energy dependence should be evaluated. If energy dependence is severe enough to warrant, efficiency factors must be modified

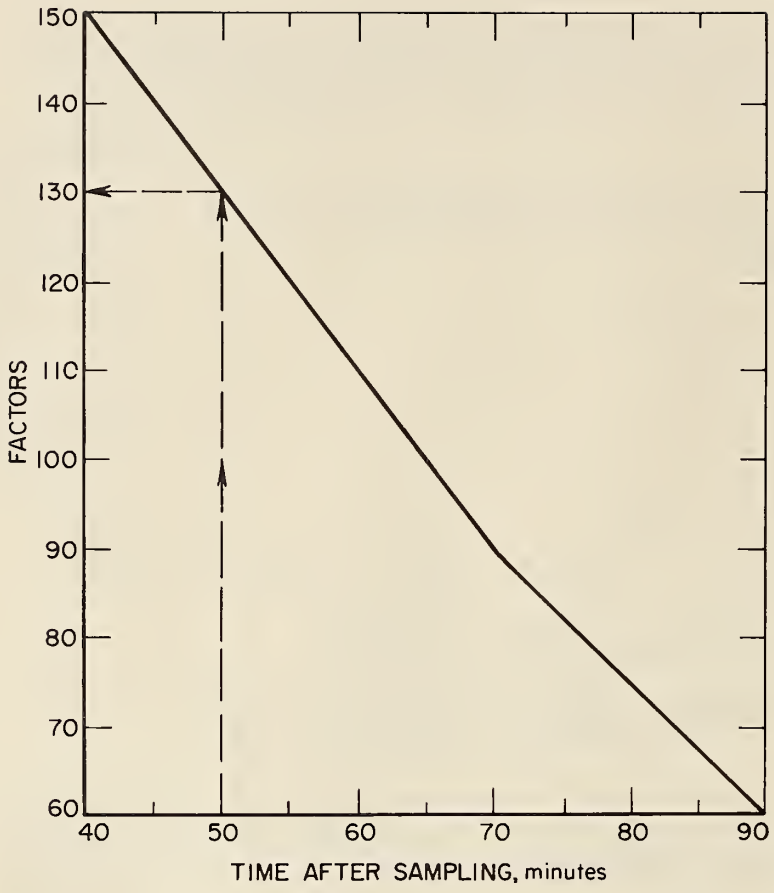

FIGURE 7. - Factors for converting disintegrations per minute per liter of air sampled, 40 to 90 minutes after sampling, to radon-daughter working levels.

accordingly. A portable digital scaler is almost essential for accurate counting of the relatively low-level activity resulting from thoron-daughter decay. Total disintegrations can be accumulated on the scaler for several minutes and then averaged for better counting statistics.

Total alpha-particle disintegrations per minute divided by the total liters of air represented by the sample indicates the dpm per liter of air. This quantity divided by the appropriate factor corresponding to the elapsed time since the end of sampling (from fig. 8) is the working level (WL) of thoron daughters present.

The technical definition for a working level of thoron (radon-220) daughters is the quantity of Pb-212 per liter of air which in decaying through Bi-212, Po-212, and Tl-208 to Pb-208 will yield 1.3×10^5/MeV of alpha energy.

Following is a sample thoron-daughter WL calculation:

Sample volume = 50 liters

Time at end of sample = 10:00 a.m.

Time of counting = 3:00 p.m.

Elapsed time = 5 hours

Factor (from fig. 8) = 13

Count rate (cpm) = 125

Counter efficiency = 25 percent

Total decay events per minute = $\frac{125}{0.25}$ = 500 dpm

WL = $\frac{500}{50 \times 13}$ = 0.77

SEPARATING RADON-DAUGHTER WORKING LEVELS FROM THORON-DAUGHTER WORKING LEVELS

The presence of radon daughters with thoron daughters should not affect the thoron-daughter working level computation, but the radon-daughter working level computation can be overestimated if appropriate correction factors are not applied. To correct the radon-daughter working level computation for alpha particle counts due to thoron-daughter decay, the thoron-daughter working level should be determined first. Where a total sample of at least 50 liters of filtered air was obtained in 5 minutes, the same filter sample can be used for both radon-daughter and thoron-daughter working level computation.

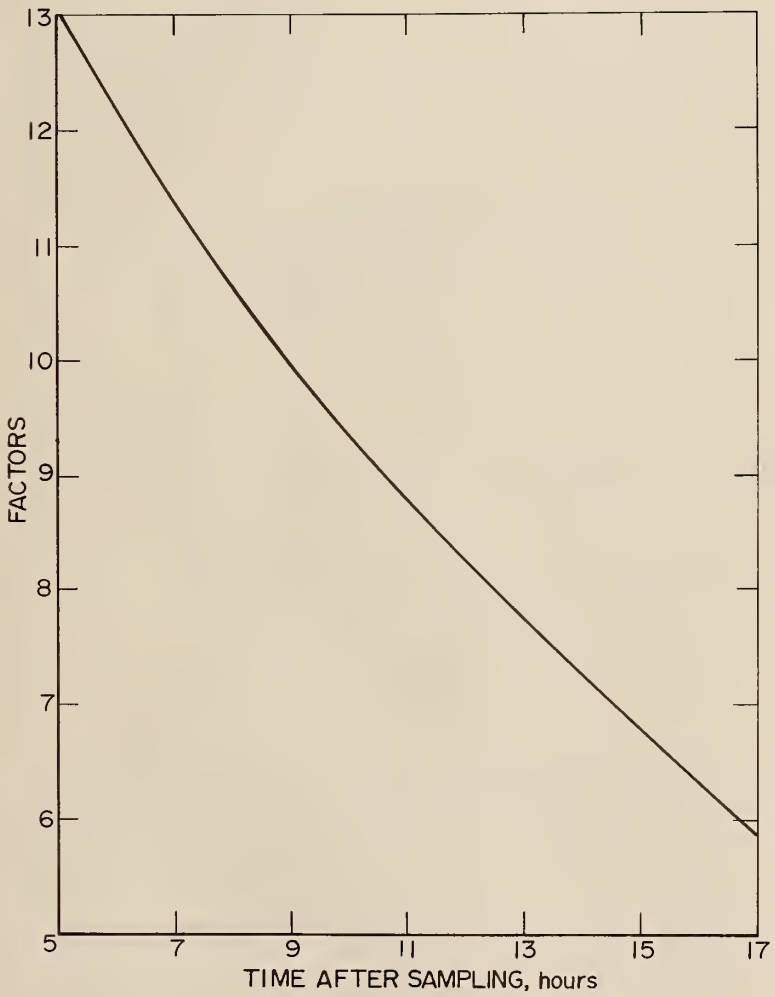

FIGURE 8. - Factors for converting disintegrations per minute per liter of air sampled, 5 to 17 hours after sampling, to thoron-daughter working levels.

In this case, the presumed radon-daughter alpha activity should be counted between 40 and 90 minutes after sampling. Counts obtained and the elapsed time between the end of sampling and counting should be recorded.

Working levels of thoron daughters are then evaluated by the previously described method.

Table 1 lists disintegrations per minute per liter of air which should be subtracted from total disintegrations per minute per liter of air, to separate thoron-daughter

activity from radon-daughter activity. Table 1 is arranged so that this activity adjustment can be made to activities obtained between 40 and 90 minutes after sampling, according to subsequently determined thoron-daughter working levels.

TABLE 1. - Dpm per liter of air which should be subtracted when calculating radon-daughter working levels mixed with thoron-daughter working levels[1]

Measured thoron-daughter working levels	Elapsed minutes after sampling when radon-daughter dpm per liter of air was counted					
	40	50	60	70	80	90
0.1	2	2	2	2	2	2
0.2	3	3	3	3	3	3
0.3	5	5	5	5	5	5
0.4	7	7	7	7	6	6
0.5	8	8	8	8	8	8
0.6	10	10	10	10	10	10
0.7	12	12	12	11	11	11
0.8	13	13	13	13	13	13
0.9	15	15	15	15	15	14
1.0	17	17	17	17	17	17
1.2	20	20	20	20	19	19
1.4	23	23	23	23	23	22
1.6	27	27	26	26	26	26
1.8	30	30	30	29	29	29
2.0	34	33	33	33	32	32

[1]Based on the assumption that ThC is nearly at equilibrium with ThB. Generally, this will result in a conservative estimate of radon-daughter working levels, especially at counting times of between 40 and 60 minutes. An underestimate of more than 10 percent should not result under the high equilibrium conditions where most screening samples will be taken. Compensation for low initial Bi-212 to Pb-212 equilibrium commences to be significant only above one working level of thoron daughters.

Working levels of thoron and radon daughters represent the same amount of alpha energy (1.3×10^5/MeV); therefore, in assessing the combined hazard, their respective working levels can be added. This is based on the assumption that the 10.6-hour half-life of Pb-212 will not affect its complete decay to Pb-208 in the lungs. Our best information to date does not allow any adjustment for the possibility that all the Pb-212 may not be retained until complete decay occurs.

Following is an example of the total working level computation from a filtered-air sample consisting of both thoron and radon daughters:

Total air volume sampled in 5 minutes = 50 liters

Time end of sample = 9:00 a.m.

Alpha activity at 9:40 a.m. = 500 cpm

Alpha activity at 2:00 p.m. = 120 cpm

Alpha scintillation detector counting efficiency = 25 percent

Thoron-Daughter Working Level Calculation

Total alpha disintegrations per minute 5 hours after sampling $= \frac{120}{0.25} = 480$ dpm

dpm/liter of air $= \frac{480}{50} = 9.6$

Decay factor for 5 hrs. (fig. 8) = 13

Thoron-daughter working level $= \frac{9.6}{13} = 0.74$

Radon-Daughter Working Level Calculation

Total alpha disintegrations per minute, 40 minutes after sampling $= \frac{500}{0.25} = 2{,}000$

dpm/liter of air $= \frac{2{,}000}{50} = 40$

dpm/liter of air from 0.74 WL of thoron daughters 40 minutes after sampling (from table 1) = 12

dpm/liter of air due to radon daughters = 40-12 = 28

Decay factor for 40 minutes (fig. 7) = 150

Radon-daughter working level $= \frac{28}{150} = 0.19$

Combined Thoron Daughter-Radon Daughter Working Level

0.74 + 0.19 = 0.93 WL

Small-volume capacity pumps will require more than 5 minutes to collect a 50-liter sample of filtered air, which is the minimum amount required to properly evaluate thoron-daughter concentrations. In this case, a separate simultaneous 5-minute radon-daughter sample will be necessary and the appropriate activity correction factor from table 1 must be applied to compute the correct radon-daughter working level.

CONTROL OF THORON DAUGHTERS

The relative half-lives of the members of the thoron- and radon-daughter decay series determine how effective ventilation is in their control. The

rule is that the longer the half-life of a particular radionuclide, the more effective ventilation is in control of the radionuclide. Of course, if precursors of short-lived radionuclides have long half-lives, their control by ventilation also improves the general control of subsequent but shorter-lived members of the decay series.

Ventilation is relatively more effective in controlling radon gas with a half-life of 3.8 days than in controlling thoron gas having a half-life of only 54.5 seconds. However, because of the relative short half-lives of Po-218, Pb-214, and Bi-214 (3.05 m, 26.8 m, and 19.7 m, respectively) compared to the half-life of Pb-212 (10.6 h), ventilation is much more effective in the relative control of thoron daughters than radon daughters. An air change every 10 minutes limits thoron-daughter potential alpha energy to about 1 percent of its maximum, while the same ventilation rate limits potential alpha energy from radon daughters to about 18 percent of its maximum. Therefore, hazardous concentrations of thoron daughters cannot exist except where thoron gas emanation rates are very high and/or ventilation rates are very low.

Bureau personnel measured thoron-daughter concentrations as high as 0.6 WL in the previously mentioned Alaskan uranium mine. In general, radon-daughter working levels were more than twice as high as thoron-daughter working levels measured. This would seem to be a lower ratio than might be expected; however, information regarding relative thoron and radon gas concentrations was not obtained.

CONCLUSION

Although thoron daughters have not been recognized as a significant health hazard in United States mines, their existence as a health hazard is a possibility. Where ores associated with thorium-232 are mined by underground methods, the hazard could be present, especially if good ventilation is lacking. The health hazard might go undetected if aerosol samples of mine air are not taken for radioactivity measurements. Another possible way to overlook a thoron-daughter health hazard would be to interpret thoron-daughter alpha activity on a filter as being due to radon daughters.

From table 1, 0.3 WL of thoron daughters is depicted by a total of only 50 alpha disintegrations per minute on a 10-liter aerosol filter-sample, counted 40 to 90 minutes after sampling. Considering the fact that most alpha detectors are about 25 percent efficient, only about 12 counts per minute would be actually registered on a count-rate meter or scaler. This amount of activity would indicate only 0.03 to 0.08 WL of radon daughters and would be very difficult to detect on the most sensitive scales of most count-rate meters. Therefore, at least 50 liters of air must be sampled when screening mines for possible health hazards due to the presence of airborne radioactive contaminants.

If radioactivity is measurable in the aerosol sample, the methods described in this paper should be used to determine the nature of the radioactivity and to analyze it relative seriousness as a health hazard.

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